Mechanical Properties of Blends of HDPE and Recycled Urea-Formaldehyde Resin

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ABSTRACT: The mechanical properties of blends of high-density polyethylene (HDPE) with a recycled thermosetting filler, urea-formaldehyde grit (UFG), were evaluated in the range of 0–23% of filler by volume. Ethylene-acrylic acid (EAA) copolymers and an ionomer based on EAA were evaluated as compatibilizers. The observed tensile modulus of the ionomer-treated blends was raised to three times the modulus of virgin polyethylene, whereas the modulus of the untreated blends reached double that of polyethylene. The ionomer-treated blends also showed a higher tensile strength than the blends without filler treatment. The improvement in the properties was assigned to an increased interaction between the filler and the polymer matrix. © 2000 John Wiley & Sons, Inc. J Appl Polym Sci 77: 3220–3227, 2000

Key words: thermoset recycling; urea formaldehyde resin; filled HDPE

INTRODUCTION

In the last decade, plastics recycling has received considerable attention amid growing environmental concerns about scrap disposal, as well as potential economic benefits of using scrap plastic.[1–4] The economic benefits stem from the scrap's low cost, which may even reach negative values due to tougher environmental restrictions on conventional disposal. Manufacturers are increasingly expected to have cradle-to-grave responsibility for the materials they produce.[3] Recycling is viewed as a partial solution, by delaying the ultimate scrapping of the polymer and, in some cases, reducing the demand for virgin resins.

The recycling of thermoset resins has suffered from the perception that they cannot be reused because, unlike thermoplastics, glass, and metals, thermosets cannot be remelted. However, in recent years, considerable progress in thermoset recycling has altered that view.[3] Some of the existing recycling technologies include the addition of regrind into thermoplastics or thermosets, the recovery of raw materials via hydrolysis or glycolysis, the recovery of chemicals via pyrolysis, and energy recovery through incineration.[1,3]

Urea-formaldehyde (UF) accounts for about 15% of the total thermoset resin production. Currently, one of its major applications is in molded products, including electrical equipment, dinnerware, buttons, cosmetic caps, and bottles. However, the same factors that make UF a good choice for many applications, namely its chemical, thermal, and mechanical stability, are also what make recycling such a big challenge.

This work describes the recycling of UF-molded products as fillers in high-density polyethylene (HDPE). Currently, most of the fillers used in thermoplastic materials are inorganic materials, such as glass beads, calcium carbonate, talc, kaolin, and various silicates. However, a major prob-

lem of such materials is the nonuniformity of the resulting properties due to poor dispersion of the filler in the low-polarity polymer matrix. That problem has traditionally been solved with the surface treatment of the dispersed phase with the help of a suitable coupling agent, or compatibilizer. The compatibilizing agent acts as a surfactant, lowering the interfacial energy and promoting interfacial adhesion between the dispersed phase and the polymer matrix. There are numerous examples in the literature concerning the effect of compatibilization.[5–9]

Using recycled thermosets as fillers should also be economically advantageous. For example, spent blast media (for paint removal and cleaning) is very inexpensive at about $-0.20/lb, as it would otherwise need to be recovered and disposed of properly for a price of about $1.00–1.50/lb. The pallet industry is currently seeking improved warehouse pallets to replace the currently used wooden ones. In response to this, the injection-molding industry is prepared to produce about 1 million pallets per year from HDPE. Adding a thermoset filler to HDPE will give a higher modulus than the virgin material, while at the same time lowering the cost of the final product because of the negative cost of the filler.

The work described here was confined to one readily available UF resin and particle size. The volume fraction range of the UF filler was limited to ~25 wt %, as above this level there were problems with mixing. EAA and EAA-based ionomers were used as compatibilizers.

Theoretical Background

The mechanical properties of two-phase composites made up of a continuous polymer phase and particulate filler phase have been studied in great detail. As a result, a variety of models are available to describe the modulus, tensile strength, and elongation at break as a function of filler volume fraction.

A simple model that has been used to investigate adhesion between a spherical filler and an incompressible matrix is the Einstein equation, which is valid only at low concentrations of filler particles. For the familiar case of perfect adhesion and isolated filler particles, Einstein's equation has the form[5,6]

$$E = E_m(1 + 2.5\phi), \tag{1}$$

where φ is the volume fraction of particles and E_m and E are the moduli of the matrix and composite,

respectively. A further development, the Guth and Smallwood equation

$$E = E_m(1 + 2.5\phi + 14.1\phi^2) \tag{2}$$

has been extensively investigated.[10,11] For the case of poor adhesion, where the polymer matrix slips by the filler particles, the form is[5,6]

$$E = E_m(1 + \phi). \tag{3}$$

Sato and Furukawa have developed an expression for the modulus for the case where the adhesion is so poor that the polymer matrix pulls away from the filler surface to give cavities around the filler particles.[12,13] Their equation is

$$E = E_m\left[\left(1 + \frac{\phi^{2/3}}{2 - 2\phi^{1/3}}\right)(1 - \psi\zeta) - \frac{\phi^{2/3}\psi\zeta}{(1 - \phi^{1/3})\phi}\right] \tag{4}$$

where

$$\psi = \left(\frac{\phi}{3}\right)\frac{1 + \phi^{1/3} - \phi^{2/3}}{1 - \phi^{1/3} + \phi^{2/3}} \tag{5}$$

and ζ is the adhesion parameter; $\zeta = 1$ for poor adhesion, and $\zeta = 0$ for perfect adhesion.

For expansible polymers incorporating rigid, spherical particles featuring some adhesion, the Kerner equation can be used to estimate the modulus.[11,14] The usual form is:

$$E = E_m\left[1 + \frac{15(1 - \nu)}{8 - 10\nu}\frac{\phi}{1 - \phi}\right] \tag{6}$$

where ν is Poisson's ratio of the matrix (for HDPE, $\nu \sim 0.35$).

In contrast with the tensile modulus, theoretical predictions of the tensile strength are less highly developed. However, extensive work has been reported by some authors, including Nielsen,[13] Piggot and Leidner,[15] and Nicolais and Narkis.[16] A characteristic of all theories is the relationship between volume fraction and projected area fraction of the particulate inclusions. Nicolais and Narkis proposed that the area fraction depends on the volume fraction to the two-thirds power,

$$\sigma = \sigma_m(1 - K\phi^{2/3}) \tag{7}$$

whereas Piggot and Leidner suggested a first-power relationship

$$\sigma = \sigma_m(1 - \phi) \tag{8}$$

where σ and σ_m are the tensile strengths of the composite and the polymer matrix, respectively. The parameter, K, in the Nicolais–Narkis model accounts for the adhesion between the filler particles and the matrix; the lower the value, the better the adhesion. The theoretical value of K for the extreme case of poor adhesion is 1.21. The Piggot–Leidner expression

$$\sigma = \sigma_m(1 - B\phi) \tag{9}$$

includes a similar parameter B that accounts for the weakness in the structure due to stress concentration.

Nielsen[17] suggested that another way of representing the tensile strength is to consider a two-phase system with poor adhesion as a matrix with voids. In this extreme, the filler occupies the voids without having any influence on the mechanical properties of the composite due to the absence of adhesion at the interfacial boundary. According to the porosity theory, which has been widely used for nonpolymeric materials such as metals and ceramics,[18] the specific change in tensile strength $d\sigma/\sigma$ is directly proportional to the porosity, P,

$$-\frac{d\sigma}{\sigma} = aP \tag{10}$$

where a is the proportionality constant, and the negative sign represents the decrease in tensile strength with an increase of porosity. Replacing the porosity with the filler volume fraction and integrating leads to the expression

$$\sigma = \sigma_m\exp(-a\phi). \tag{11}$$

The parameter a is suggested[18,19] to be related to stress concentration; the higher the value of a the greater the stress concentration effect or the poorer the adhesion.

Turcsanyi et al. used the simple empirical model

$$\sigma = \frac{1 - \phi}{1 + A\phi}\,\sigma_m f(\phi) \tag{12}$$

to describe the composition dependence of the tensile strength.[19,20] The value of A depends on the type of packing of the filler particles. The value is 2.427 for a hexagonal close packing and 2.318 for face-centered cubic packing. A value of $A = 2.5$ was assumed as an approximate upper limit. The authors also explored the dependence of σ_p on volume fraction, which, after applying eq. (12) to a number of different polymer/filler systems, was best described by an exponential function. The resulting expression was

$$\sigma = \sigma_m\frac{1 - \phi}{1 + 2.5\phi}\,\exp(B\phi) \tag{13}$$

where B is a parameter related to the interfacial properties. For example, $B = 0.246$ for an ABS polymer filled with glass beads, approximating the no-adhesion case. Surface treatment to improve the adhesion between the phases increased B to 1.059.

The decrease in elongation at break in filled-polymer composites is due to the fact that the deformation of the filler is generally much less than that of the polymer matrix; thus, the filler forces the matrix to deform more than the overall deformation of the composite. A basic model that describes the elongation at break was developed by Nielsen. For the case of perfect adhesion, under the assumption that the polymer breaks at the same elongation in the filled system as in the neat polymer, the elongation at break is given by

$$\varepsilon = \varepsilon_m(1 - \phi^{1/3}) \tag{14}$$

where ϵ and ϵ_m are the elongations at break of the composite and the unfilled polymer, respectively.[13] In the case of poor adhesion, the elongation is expected to decrease more gradually than in the case of perfect adhesion. The poor-adhesion case can be derived[13] by combining the Sato–Furukawa modulus model for no adhesion and the Nicolais–Narkis tensile strength model with $K = 1.21$.

EXPERIMENTAL

Materials

The urea-formaldehyde grit (UFG) for this study was provided by Composition Materials Co. of Fairfield, CT (www.compomat.com). The particle size of the UFG was 110–240 μm, based on area. A density of 1.5 g/cm^3 was used to calculate the

volume fractions of filler. The compatibilizers were ethylene-acrylic acid copolymers (EAA; Scientific Polymer Products Inc., www.scientificpolymer.com) and sodium and zinc ethylene-acrylic acid ionomers (Exxon). The EAA contained 5, 10, or 15% acrylic acid. The zinc ionomer (Iotek 7010) was based on 15% EAA with 35 mol % of the acrylic acid groups neutralized to the zinc salt. The sodium ionomer (Iotek 8000) was 15% EAA with 45 mol % of the acrylic acid groups neutralized to the sodium salt. These will be referred to as Zn ionomer and Na ionomer, respectively. For the matrix, two very similar HDPEs were used. For series with the EAA compatibilizer, the resin was a Union Carbide (www.unioncarbide.com) DMDA-1200 NT P with a specified melt index of 0.7 g/10 min and a density of 0.959 g/cm^3. The second, a virgin HDPE resin (Phillips Petroleum, EHM 6007, www.resinnet.com) with a density of 0.964 g/cm^3 and a melt index of 0.75 g/10 min (190°C, 2.16 kg), was used with the ionomer series.

Blend Preparation

The properties of the blends were studied as a function of the amount of UFG filler. The UFG was first preheated to about 100°C for 30 min and coated with a compatibilizer by intensive mixing in a blender for 1 min. The amount of compatibilizer was fixed at 10 wt % of the UFG, except for untreated UFG. The UFG aliquots were then dry-blended with HDPE pellets and extruded by using a 1-in. single-screw extruder (Wayne Machine and Die Co., www.waynemachine.com) featuring a Maddock mixing section. The temperatures in all three zones of the extruder and the die exit were set at 200°C, and the speed was set at 80 rpm. The extrudate was fed directly into a Rancastle Extrusion Systems Inc. pelletizer to prepare the blends for injection molding. Blends of 0, 5, 10, 15, 20, and 25 wt % of UFG were usually prepared. In all cases, the 0% UFG blend was treated exactly the same as the blends of higher concentration. The mixes featuring the EAA-15 were abandoned, because of the extremely sticky nature of the blends, suggesting exudation of the compatibilizer from the UFG surface. Tensile specimens were molded into dumbbell shape using a 1-ton Arburg model 221–75-350 injection-molding machine.

Tensile Testing

Tensile specimens were tested on an Instron 1101 according to the ASTM D638M test procedure.
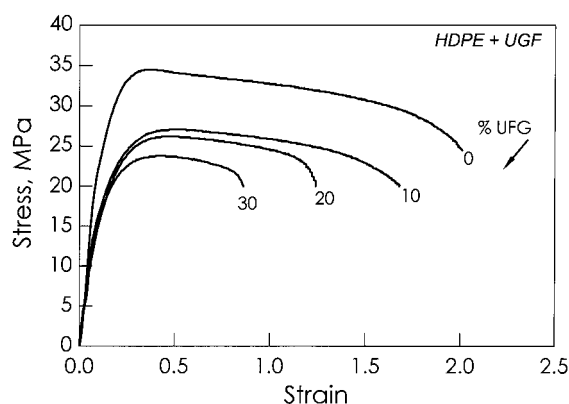

Figure 1 Stress–strain behavior of UFG-filled HDPE at various filler levels (wt %).

Testing was done at room temperature by using a crosshead speed of 50 mm/min and a load range of 0–500N. The mean value of at least eight specimens of each sample was taken, although specimens that broke in an unusual manner were disregarded.

Adhesive Testing

An adhesion test, modeled after the ASTM D903–93 procedure, was used to test the peel strength of the adhesive bond between UFG and HDPE with the EAA copolymers as the adhesives. The surfaces of both the UFG and the poly(ethylene) (PE) tape were wiped clean with acetone before bonding. The test specimens were prepared in a heat press at a temperature of 100°C, assuring that an ~0.22-mm EAA bonding layer remained between the two adherents. The samples were tested at room temperature on an Instron 1101, by using a crosshead speed of 20 mm/min and a load range of 0–50N. At least half the bonded area of the specimens was peeled. The peel force was determined by taking the average of the load curve, using the data after steady state had been reached. The peel strength, defined as the peel force per unit width of bond line, was calculated in newtons per meter from the resulting peel force and the strip width (~1 cm).

RESULTS AND DISCUSSION

To gain some basic knowledge of the HDPE/UFG system, the stress–strain behavior was examined by using blends with 0, 10, 20, and 30 wt % filler. The stress–strain results are given in Figure 1. As
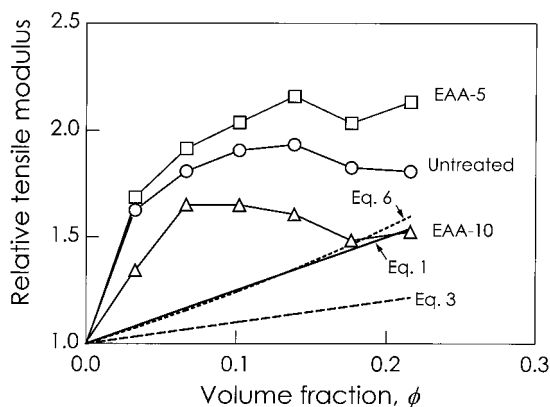
Figure 2 Influence of EAA modification on the relative tensile modulus of HDPE/UFG (ratio of EAA/UFG was held constant at 1 to 10). The Einstein equations for perfect and poor adhesion, and the Kerner equation are shown for comparison.

the concentration of UFG was increased, both the tensile strength and the elongation at break decreased. Because of the scale of the figure, the substantial increase in the Young's modulus (slope at zero strain) is not evident.

Figure 2 shows the effect of EAA modification of the UFG filler on the relative tensile modulus of the system. At low-volume fractions (up to about 0.08), the observed modulus climbed more rapidly than predicted by any of the models discussed above, even in the absence of the EAA. For higher volume fractions (up to about 0.23), the modulus increased only slightly, as would be predicted by Einstein's equation for poor adhesion. As can be seen, the rapid increase at low-volume fractions depends heavily on the first data point, which was compensated somewhat by calculating the relative modulus by using extrapolations through all the data points and by using the intercept for the reduction (i.e., not the first observation). The extrapolating function in all cases was $E = \alpha_1(1 + \alpha_2 \phi^{\alpha_3})$, where α_1 is the desired intercept. This procedure was used to emphasize the changes in modulus and the comparison with the theory. To explain the steep increase at low-filler fractions, one could argue that the first small amounts of UFG would concentrate in the amorphous fraction and have a highly exaggerated effect on the overall modulus. Studies[21] of the influence of filler on the HDPE crystallinity suggest a slight increase of crystallinity, in addition. For particle contents greater than $\phi = 0.08$, the filler inclusions undoubtedly form aggregates. If the bond between the filler particles is not as

strong as that between the filler and the matrix, cavities would easily form. Plotted for comparison are the Einstein equation with and without adhesion, and the Kerner equation. None of these equations can describe the observations.

Although the nonmonotonicity of the modulus with acrylic acid (AA) content in the EAA (Fig. 2) could well be spurious, it also is consistent with a compatibilizing effect at low AA, followed by a softening effect on the matrix at high AA content.

The results of the tensile strength tests are shown in Figure 3. Filler treatment with both EAA-5 and EAA-10 had no significant effect on the properties, which implies that AA content has little influence on the interfacial adhesion between the filler and the matrix in this system. To explore more carefully the degree of interfacial adhesion between the two phases, the results were analyzed by using models featuring an adhesion parameter. For example, describing the entire data set with the Nicolais–Narkis model gives a $K = 0.62 \pm 0.07$. A value lower than 1.21 indicates that even without the EAA, there is significant adhesion between the UFG and the polymer matrix, according to this analysis. The same picture emerged with the model of Turcsanyi et al. [eq. (13)]. The value of $B = 2.2 \pm 0.2$ resulted when this equation was used with the pooled strength data, which is indicative of good adhesion.

The elongations at break are shown in Figure 4. Similar to the tensile strength, the elongation at break was observed to decrease gradually with an increase in the filler volume fraction. EAA treatment also did not affect significantly the
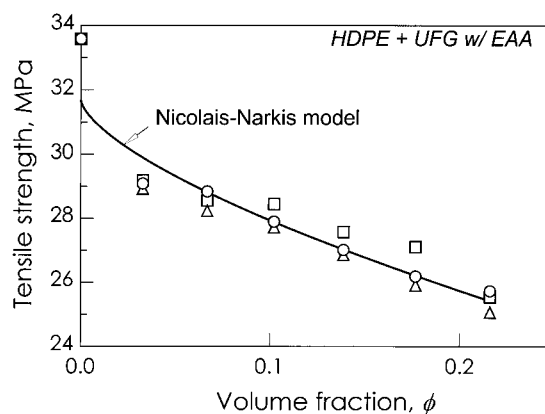

Figure 3 Tensile strength results for HDPE/UFG blends with EAA compatibilizer. The symbols are the same as for Figure 2. The Nicolais–Narkis model has been fitted to the pooled data.
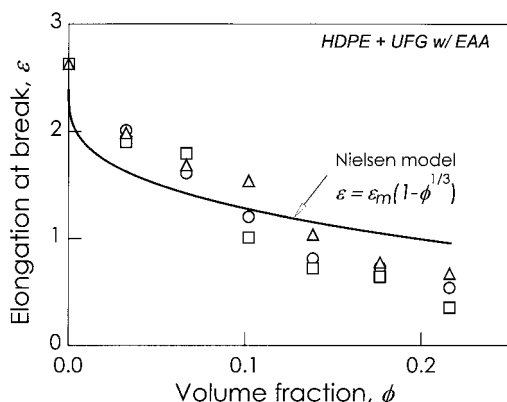
Figure 4 Elongation at break for HDPE/UFG blends with EAA compatibilizer. The symbols are the same as for Figure 2. The Nielsen model [eq. (14)] is shown for comparison.

Figure 6 Influence of ionomer modification on the relative tensile modulus of UFG-filled HDPE. The ratio of ionomer to UFG was held constant at 1 to 10. The Einstein equation is shown for comparison.

elongation at break. The data is compared with Nielsen's equation for good adhesion.

Although the tensile results are not all independent, they do reflect different degrees of deformation. The surprising result is that they all indicate some adhesion of even untreated UFG to HDPE.

Figure 5 depicts the peel strength as a function of AA content in an EAA adhesive layer. The highest values of peel strength were achieved with EAA-15 as the adhesive: 200–300 N/m compared to 150–200 N/m for both EAA-5 and EAA-10. The type of failure between the two members in all specimens was cohesive, as the failure occurred in the EAA adhesive itself. This suggests that EAA should be effective in compatibilizing the two phases of the composite. Using a LLDPE
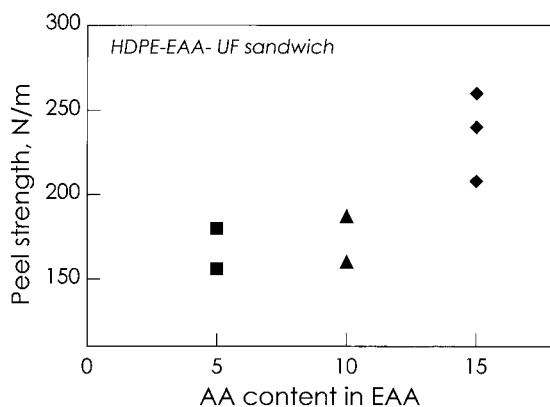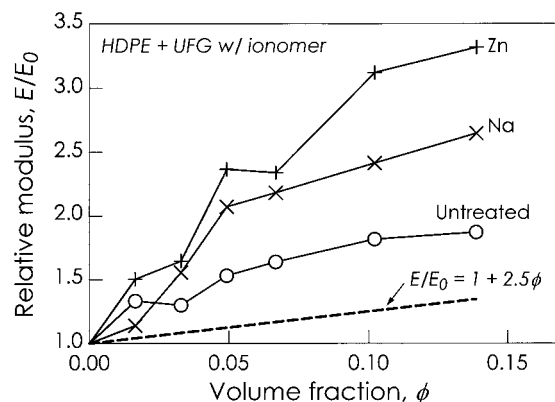

Figure 5 Effect of AA content of the EAA adhesive layer on the peel strength of HDPE/EAA/UF sandwiches.

(0% AA) as the adhesive, resulted in peel forces that were too low to measure accurately.

The effects of ionomer modification of the UFG on the relative tensile modulus of the blends are shown in Figure 6. The results suggest that the ionomer treatments led to a significant improvement in the modulus compared to the untreated blend. As was found with the EAA series, the experimental data of the untreated blend exceeds somewhat the Einstein's equation for good adhesion. The results for both ionomer treatments, however, are far above the untreated blend. Both sets of data were also compared with the values calculated from Kerner's equation; this model describes satisfactorily the modulus of the untreated blends, but the data for the blends with the ionomers, especially the zinc, were poorly represented, suggesting that their behavior is quite unusual. Compared to the effect of EAA modification, treatment of the UFG filler with either ionomer led to a more significant increase in the tensile modulus. The incorporation of the zinc ionomer and 20 wt % of UFG filler increased the modulus of virgin HDPE by a factor of 3, whereas the same EAA and filler content only doubled the modulus.

Putting the modulus values into perspective is important. At a 15 vol % filler level, the relative changes in modulus observed in this work varied from about 1.5 to 3.5, depending on the amount and type of compatibilizer. These values are comparable to similar PE systems using typical inorganic fillers. For example, some reported relative modulus values are about 2.0 with wollastinite,[14] 1.5–1.8 with $CaCO_3$,[22–23] 1.5 with aluminum,[24]
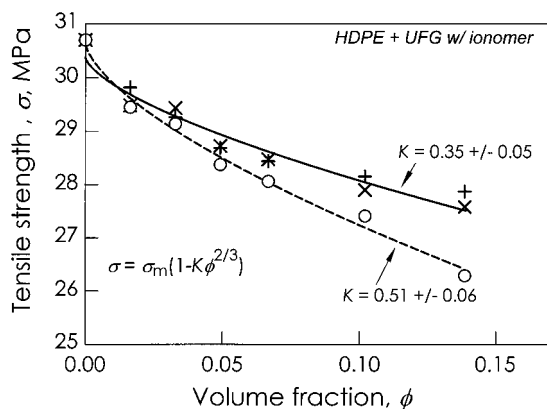
Figure 7 Tensile strength results for the ionomer-modified blends. The symbols are the same as in Figure 6.
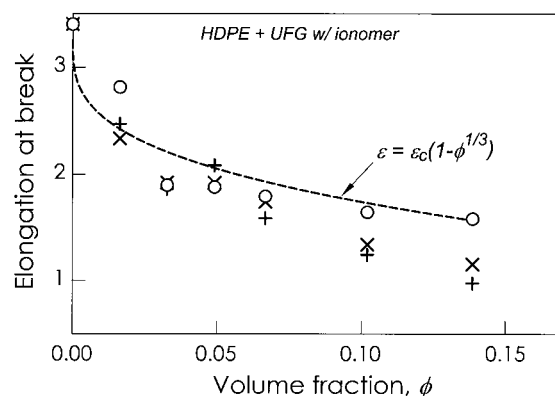

Figure 8 Elongation at break for the blends of the ionomer-modified blends. The symbols are the same as in Figure 6. Shown is the Nielsen equation fitted to the data for the unmodified blends.

1.6 with kaolin,[23] 1.3 with talc,[25] and 1.5 with mica.[25] The higher modulus of the inorganic fillers plays a minor role with the relatively low modulus matrix.

The tensile strength results for the ionomer composites are shown in Figure 7. In contrast to the EAA results, the blends with ionomer-treated filler showed higher tensile strengths than the blends with untreated filler. Describing the untreated blends data with the Nicolais–Narkis theory resulted in $K = 0.51 \pm 0.06$, whereas the treatment with Zn ionomer gave $K = 0.38 \pm 0.09$; pooling the data for the two ionomers yielded $K = 0.35 \pm 0.05$, as shown in Figure 7. Both treated and untreated blends gave K's that are lower than 1.21, which indicate good adhesion between the UFG and the polymer matrix. The experimental

data were also compared with both the porosity model [eq. (11)] and the Turcsanyi model [eq. (13)]. These two models also indicate good interfacial adhesion between the filler and the matrix, even for the untreated blends. The parameters of all three theoretical models are assembled in Table I. Based on the standard error of estimates, the fits for the Nicolais–Narkis model appear to be slightly better than those for the other two-parameter models.

The elongation at break results are shown in Figure 8. The values for the ionomer-treated blends are somewhat lower than the values for the untreated blends. This is another indication that ionomer treatment of the UFG improves interfacial ad-

Table I   Parameters Found for Fits of Two-Parameter Tensile-Strength Models to Data for Ionomer-Modified HDPE/UFG Blends

| Model | Additive | σ_m (MPa) | Adhesion Parameter | | | SEE |
			Symbol	Found	No-Adhesion Value	
Nicolais–Narkis	None	30.6 ± 0.3	K	0.51 ± 0.06	1.21	0.072
[eq. (7)]	Na ionomer	30.4 ± 0.5		0.35 ± 0.09	(Lower indicates	0.114
	Zn ionomer	30.5 ± 0.4		0.38 ± 0.08	better adhesion)	0.095
Porosity	None	30.2 ± 0.5	a	1.03 ± 0.25	Lower indicates	0.20
[eq. (11)]	Na ionomer	30.1 ± 0.9		0.66 ± 0.32	better adhesion	0.19
	Zn ionomer	30.1 ± 0.6		0.72 ± 0.33		0.17
Turcscanyi et al.	None	30.3 ± 0.4	B	2.20 ± 0.20	~ 0.25	0.11
[eq. (13)]	Na ionomer	30.3 ± 0.5		2.36 ± 0.29	(Higher indicates	0.14
	Zn ionomer	30.3 ± 0.5		2.49 ± 0.22	better adhesion)	0.13

The ± terms are 95% confidence intervals. SEE is the standard error of estimate.

hesion between the filler and the matrix. The data in this figure are compared with Nielsen's model for good adhesion between filler and matrix. Note that the Zn ionomer treatment produced higher modulus values than Na ionomer, whereas the differences in both the tensile strength and the elongation at break are not significant. Thus in going from sodium to zinc neutralization, there appears to be a gain in modulus without a corresponding loss in elongation, an unusual occurrence for blends of this type. The elongations found with the HDPE/UFG blends are slightly lower than those reported for clay- and $CaCO_3$-filled MDPE.[23]

CONCLUSION

UFG fillers can be used effectively in blends with HDPE, as one of the possible applications for this recycled thermoset. When compared with more traditional fillers, the UFG is lighter and less expensive, and the modulus gains are similar, although the UFG-based systems are likely to be more brittle. The results of the present study suggest that the addition of zinc-neutralized ionomer can produce significant increases in modulus, at only a modest cost in elongation to break.

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